Time Reversal Symmetry Breaking and Spontaneous Currents in s-Wave / Normal Metal / d-Wave Superconductor Sandwiches

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We study the physical properties of an *s*-wave – normal metal – *d*-wave junction in terms of the Andreev bound states in the normal metal layer. The phase dependence of bound states with different orientations leads to superconducting states with broken time reversal symmetry for generic orientations of the *d*-wave superconductor crystal. The occurrence of such a state and the associated spontaneous supercurrent along the junction is analyzed in the framework of Ginzburg-Landau theory and by the solution of the Bogoliubov-de Gennes equations.

During the last few years the order parameter (OP) symmetry has been one of the intensively debated issues in the field of high-temperature superconductivity. A growing number of experiments leaves little doubt that the basic symmetry of the Cooper pairs has $d_{x^2-y^2}$-wave character in many of the high-temperature superconductors (HTSC) [1,2]. The unconventional symmetry of the OP has important implications for the Josephson effect. For d-wave superconductors the Josephson coupling is subject to an additional phase dependence caused by the internal phase structure of the Cooper pair wave function. The phase properties of the Josephson effect have been discussed within the framework of the generalized Ginzburg-Landau (GL) [3] as well as the tunneling Hamiltonian approach [4]. It was found that the current-phase relation depends on the mutual orientation of the two coupled superconductors and their interface. This property is the basis of all the phase sensitive experiments probing the OP symmetry. In particular, it is possible to create multiply connected d-wave superconductors which generate half-integer flux quanta as observed in experiments [5].

Various interesting phenomena occur in 45°-interfaces of $d_{x^2-y^2}$-wave superconductors, where one of the nodes of the pair wave function lies parallel to the interface normal vector (Fig. 1). For an interface to a normal metal or an insulator a bound state appears at zero energy giving rise to a zero-bias anomaly in the I-V-characteristics of quasiparticle tunneling [6,7]. It was also shown that in such an interface to an s-wave superconductor the energy minimum corresponds to a Josephson phase different from 0 or π [8]. Based on Ginzburg-Landau theories it was suggested that this is connected with a local breakdown of time reversal symmetry $\mathcal{T}$ [9,10]. The s-wave and d-wave OP can form a complex combination, a so-called $(s+id)$-state, close to this 45°-junction. This leads to a phase difference of $+\pi/2$ or $-\pi/2$ across the interface, which corresponds to two degenerate states [10,11]. It can be seen from the GL formulation that under this condition a spontaneous current flows parallel to the in-

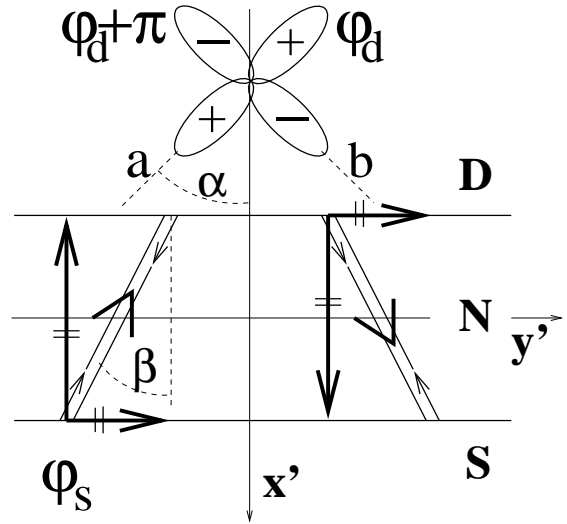terface which produces a local field distribution [9].

FIG. 1. Schematic view of the SND-junction. The angle α denotes the orientation of the d-wave superconductor (crystal axis a and b) and β the momentum direction of the bound state. The currents generated by the bound states tend to cancel in the direction perpendicular to the interface, whereas they add parallel to the interface to a spontaneous current.

In this paper we consider a 45°-interface with a normal metal between the d-wave and the s-wave superconductor, a device which we call the SND-junction [12]. Also for this configuration a $\mathcal{T}$-violating state appears and generates a supercurrent mainly in the region of the normal metal. It is our goal to demonstrate that this current has a simple and intuitive interpretation in terms of subgap Andreev bound states in the sandwiched normal metal layer. Let us first outline the basic idea for the situation shown in Fig. 1 where $\alpha = \pi/4$ and the c-axis is parallel to the interface. In terms of the phase difference $\varphi = \varphi_d - \varphi_s$, the Josephson current carried by a bound state with a specific orientation β can be expanded in harmonics of the phase difference

φ as $I_\beta(\varphi) = I_1(\beta)\sin(\varphi) + I_2(\beta)\sin(2\varphi) + \cdots$. In the geometry considered, each bound state with orientation $0 < \beta < \pi/4$ that sees the "+" lobe with phase φ_d, has a mirror bound state with orientation $-\beta$ that sees the "–" lobe of the $d_{x^2-y^2}$-pair wave function with phase $\varphi_d + \pi$. As a result, in the total current perpendicular to the interface, all odd harmonics cancel, and the Josephson coupling is reduced. The leading term is $I_\perp \sim \sin(2\varphi)$ [13] and the stable ground state with $I_\perp = 0$ is at $\varphi = \pm\pi/2$ and, thus, breaks time reversal symmetry. The Josephson current parallel to the interface, however, has contributions from the odd harmonics and to leading order $I_\parallel \sim \sin(\varphi)$. Remarkably, this parallel contribution is *nonzero* in the ground state and constitutes a spontaneous current.

Let us first consider this property of the SND junction on a phenomenological level by means of GL theory. We describe the superconducting state by two OP's, η_s (s-wave) and η_d (d-wave), which correspond to the local pairing amplitudes. The corresponding GL free energy $\mathcal{F}$ has the general form,

$$\frac{\mathcal{F}}{f_0} = \int d^3r \Big[\sum_{\mu=s,d} \{ (\frac{T}{T_{c\mu}} - 1)|\eta_\mu|^2 + \beta_\mu|\eta_\mu|^4 + \xi_\mu^2|\mathbf{\Pi}\eta_\mu|^2 \}$$
$$+ \gamma_1|\eta_s|^2|\eta_d|^2 + \frac{\gamma_2}{2}(\eta_s^{*2}\eta_d^2 + \eta_s^2\eta_d^{*2}) + \frac{(\nabla\times\mathbf{A})^2}{8\pi f_0}$$
$$+ \tilde{\xi}^2((\Pi_x\eta_s)^*(\Pi_x\eta_d) - (\Pi_y\eta_s)^*(\Pi_y\eta_d) + \text{c.c.}) \Big], \quad (1)$$

where f_0 is a free energy density, T_{cs} and T_{cd} are the transition temperatures of η_s and η_d, respectively, and $\beta_{s,d}, \gamma_{1,2}, \xi_{s,d}$, and $\tilde{\xi}$ are real coefficients ($\xi_{s,d}$ corresponds to the zero-temperature coherence length). These coefficients and the transition temperatures are in general different in the three regions of the SND-junction. We use $\mathbf{\Pi} = \nabla - (2\pi i/\Phi_0)\mathbf{A}$, with vector potential $\mathbf{A}$ and flux quantum $\Phi_0 = hc/2e$. To study the properties of the SND-junction we minimize this free energy with respect to $\eta_{s,d}$ and $\mathbf{A}$. Assuming homogeneity along the interface the problem reduces to one spatial dimension which corresponds to the [1,1,0]-direction in the coordinates used in $\mathcal{F}$ ($\hat{x} = \hat{a}$ and $\hat{y} = \hat{b}$). We call this direction x' and the perpendicular ones y' and z.

We solve the complete set of GL equations numerically, for the case in which the coefficients in $\mathcal{F}$ are identical for both OP's and throughout the system. The transition temperatures are only different from zero in the corresponding superconducting regions. We assume the interfaces between the different layers to be completely transparent, i.e. the OP's are continuous and have a continuous derivative. For our calculation we choose $\beta_s = \beta_d = 1/2$, $\xi_s = \xi_d = 1$ (unit of length), $\gamma_1 = 4/5$, $\gamma_2 = 2/5$ and $\tilde{\xi} = 1$. This leads to $f_0 = H_c^2/8\pi$, where H_c is the thermodynamic critical field at $T = 0$. We fix $\Phi_0/2\sqrt{2}\pi H_c\xi_s^2 = 4$ which corresponds to the London penetration depth λ at $T = 0$ in units of ξ_s. The result
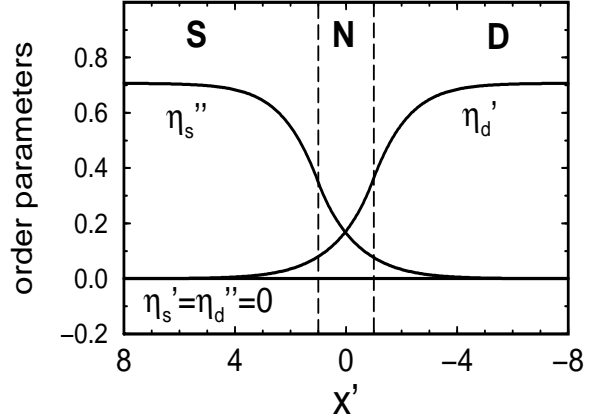

FIG. 2. Spatial dependence of the OP in the SND-junction ($\alpha = \pi/4$) based on the GL theory. The parameters of GL free energy are given in the text. The temperature is $T = T_{cs,d}/2$ and the width L of the normal metal layer is 2 in units of ξ_s.

is shown in Fig. 2 for the OP's and in Fig. 3 for the magnetic field and the supercurrent along the y'-direction.

Both OP components penetrate the normal metal layer (proximity effect) and coexist there in a combination, which for the case $\alpha = \pi/4$ is entirely determined by the mixing terms $(\gamma_2/2)(\eta_s^{*2}\eta_d^2 + \eta_s^2\eta_d^{*2})$. Within the weak coupling approach which we assume to apply, at least, within the normal metal layer, γ_2 is positive [14]. This term yields the basic $\cos(2\varphi)$-dependence of the SND-junction free energy. It fixes the phase difference between η_s and η_d to $\varphi = \varphi_d - \varphi_s = \pm\pi/2$ in accordance with the argument given above. The mixed state has the $\mathcal{T}$-violating $s \pm id$-character in the normal metal.

The supercurrent density follows from $\mathcal{F}$ as $\mathbf{J} = -2c\partial\mathcal{F}/\partial\mathbf{A}$. We find that the current component $J_{x'} = J_\perp$ vanishes in the stable junction state and that a spontaneous supercurrent flows parallel to the y'-direction and generates a magnetic field distribution B_z in and close to the metal layer (Fig. 3). Within the GL-formulation the supercurrent $J_{y'} = J_\parallel$ is caused by the spatial variation of the two OP components,

$$J_{y'} = \frac{\pi c\tilde{\xi}^2}{\Phi_0}\text{Im}\{\eta_s\partial_{x'}\eta_d^* + \eta_d\partial_{x'}\eta_s^*\}, \quad (2)$$

where we have omitted the diamagnetic part. Note that this part of $J_{y'}$ has essentially the $\sin\varphi$-dependence anticipated above. Under symmetric conditions, $J_{y'}$ depends only weakly on x' inside the normal metal layer as shown in Fig. 3. The induced magnetic field is screened perpendicular to the interface on the scale of the London penetration depth in the superconducting regions by currents flowing in the opposite direction.

Let us turn now to the microscopic view by considering the bound state solutions to the Bogoliubov-de Gennes equation in the normal metal layer [15] under the sym-
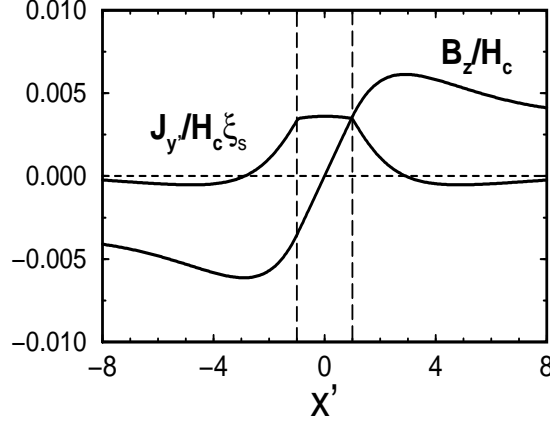
FIG. 3. Spatial dependence of the spontaneous supercurrent and the magnetic field in the SND junction based on the GL theory under the same conditions as in Fig. 2

metric condition, i.e. the d-wave energy gap in D has the form $\Delta_d = |\Delta|\mathrm{sign}(\cos[2(\theta-\alpha)])$, with the amplitude $|\Delta|$ equal to that of the gap of the s-wave superconductor in S. We take the Fermi momenta in S, N, and D to be equal and the transparency of the interfaces to be high. Furthermore, we also neglect the suppression of the energy gap near the normal metal and assume the pairing interaction to be zero in N.
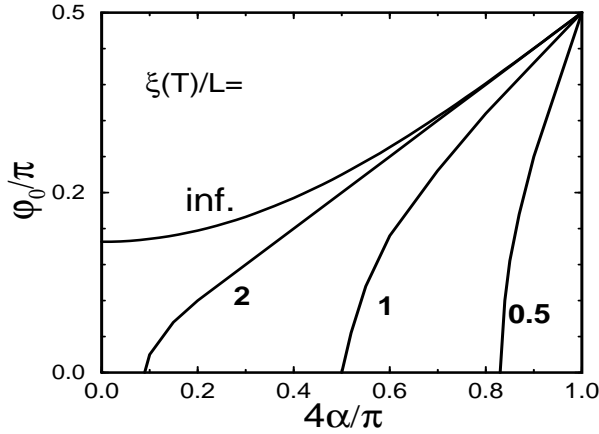

FIG. 4. The ground state phase difference $\pm\varphi_0$ as a function of orientation angle α for temperatures corresponding to $\xi(T)/L = \infty$, 2, 1, and 0.5 .

The total Josephson current is a sum over all possible (bound) states near the Fermi energy. If the width of the normal metal L is smaller than the thermal coherence length ξ_T and the elastic mean free path l in N, the Josephson current is given by [16],

$$\mathbf{J} = \int dk_y dk_z \frac{2e\mathbf{k}_F}{m\pi L} \sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{n} f_{n,\hat{\mathbf{k}}_F} \sin[n\varphi_{\hat{\mathbf{k}}_F}] , \quad (3)$$

with the Fermi momentum $\mathbf{k}_F = (k_x, k_y, k_z)$ and $k_x^2 + k_y^2 + k_z^2 \approx k_F^2$. The integral runs over all transverse momenta $|k_y|, |k_z| \leq k_F$ and the sum over all possible numbers of multiple Andreev reflections n. The factors $f_{n,\hat{\mathbf{k}}_F}$ take the suppression due to thermal decoherence and impurity scattering into account,

$$f_{n,\hat{\mathbf{k}}_F} = \exp(-2nL_{\hat{\mathbf{k}}_F}/l) \frac{nL_{\hat{\mathbf{k}}_F}/\xi_T}{\sinh[nL_{\hat{\mathbf{k}}_F}/\xi_T]} , \quad (4)$$

where we have introduced the normal metal coherence length $\xi_T = \hbar v_F/(2\pi k_B T)$ in the clean limit, the mean free path l, and the effective thickness of the normal metal layer $L_{\hat{\mathbf{k}}_F} = Lk_F/(k_F^2 - k_y^2 - k_z^2)^{1/2}$.
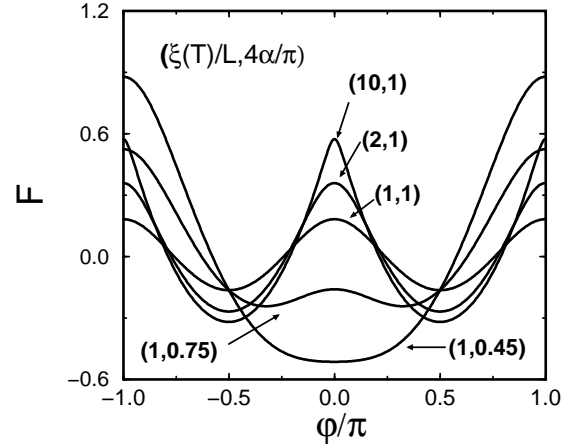

FIG. 5. The junction free energy F as a function of the phase for $\alpha = \pi/4$ and $\xi_T/L = 10, 2, 1$, and for $\alpha = 0.45\,\pi/4$ (one minimum) and $0.75\pi/4$ (two shallow minima) at $\xi_T/L = 1$.

The simplest case is that of zero temperature in the absence of impurities, so that $\xi_T = l = \infty$ and all $f_n \equiv 1$. In this limit the sums over n give sawtooth functions of the phase difference, $\mathrm{saw}[\varphi_{\hat{\mathbf{k}}_F}] = [\varphi_{\hat{\mathbf{k}}_F} + \pi]\mathrm{mod}2\pi$. We obtain the Josephson currents perpendicular and parallel to the junction immediately by angular integration,

$$I_\perp = A_\perp J_0[(\frac{\pi}{2} + \cos(2\alpha))\mathrm{saw}(\varphi)$$
$$+ (\frac{\pi}{2} - \cos(2\alpha))\mathrm{saw}(\varphi + \pi)] \quad (5)$$
$$I_\parallel = A_\parallel J_0 \sin(2\alpha)[-\mathrm{saw}(\varphi) + \mathrm{saw}(\varphi + \pi)] . \quad (6)$$

Here $A_\perp$ and $A_\parallel$ denote the perpendicular and parallel cross-section of the junction, and $J_0 = ek_F^3/(\pi mL)$. Note that the current density J_0 is inversely proportional to L, as in the GL calculation. The junction free energy $F(\varphi)$ is found by integrating $I_\perp$ with respect to the phase. It has two degenerate minima at phase differences $\varphi_0 = \pm[\pi/2 - \cos(2\alpha)]$, which correspond to a parallel current along the junction $I_\parallel = \pm B J_0 \sin(2\alpha)$. The ground state has $(s + e^{i\varphi_0}d)$-character in the normal metal layer as in the phenomenological treatment, again reflecting $\mathcal{T}$-violation.
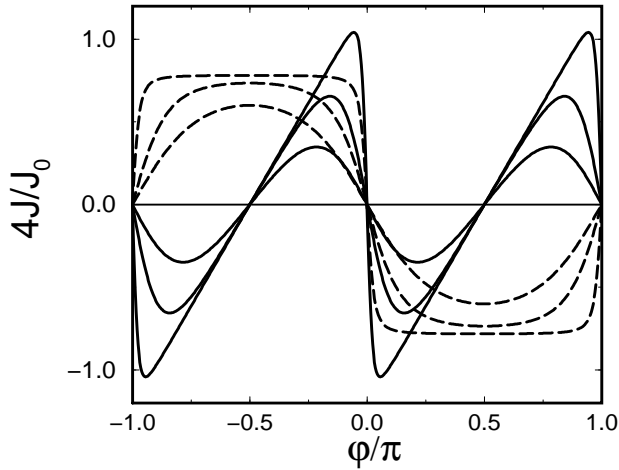
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FIG. 6. The parallel (dashed line) and perpendicular (solid line) Josephson current densities as a function of phase difference for $\alpha = \pi/4$ and temperatures corresponding to $\xi_T/L = 10, 2, 1$ (decreasing amplitude).

For nonzero temperature and in the presence of impurities, we evaluate $I_\perp$, $I_\parallel$, and the junction free energy F numerically. In Fig. 4 the equilibrium phase difference φ_0 across the junction is plotted as a function of orientation angle α for different temperatures ξ_T/L in the case $l = \infty$. We find that time reversal symmetry is broken ($\varphi_0 \neq 0, \pm\pi$) only for low enough temperatures, or for the orientation angle α exceeding a critical value. For $\alpha = \pi/4$, however, $\varphi_0 = \pm\pi/2$ for all $T < T_{cs}, T_{cd}$ as in the GL treatment. The resulting phase diagram is completely consistent with the one found by GL theories [10]. The result for the junction free energy F is plotted in Fig. 5, and $I_\perp$ and $I_\parallel$ in Fig. 6. Both temperature and disorder smear the sharp sawtooth structures found at $T = 0$ in the clean limit in a similar fashion.

The arbitrary equilibrium phase difference leads to experimentally observable effects. $\mathcal{T}$-violating junctions can lead to phase windings which are non-integer multiples of π, giving rise to non-standard (not (half-) integer) flux quantization. Thus, it is possible to create devices including $\mathcal{T}$-violating junctions which generate a spontaneous arbitrary magnetic flux [9,10]. The observation of such a deviation from standard flux quantization is a clear sign of $\mathcal{T}$-violation. Furthermore, the presence of two degenerate equilibrium states allows for hysteresis effects ($\varphi_0 \leftrightarrow -\varphi_0$). By applying a current through the junction one can switch between the two states. This effect corresponds to a phase slip with a fractional flux moving along the junction [10]. This leads to dissipation and the enhancement of microwave absorption as soon as the junction enters the $\mathcal{T}$-violating phase. The direct observation of the spontaneous currents $I_\parallel$ or the field might be difficult, since they average to zero over rather small length scales (London penetration depth). Thus a probe with high spatial resolution would be needed.

In summary, we have demonstrated that the Andreev bound states in the normal metal layer of an SND-junction are the microscopic realization of local $\mathcal{T}$-violation and provide a clear understanding of the spontaneous current found in the phenomenological Ginzburg-Landau analysis. This observation allows for a more quantitative consideration of this effect, which will be important for future experimental investigations. The experiments discussed at the end are two among several possibilities to observe this $\mathcal{T}$-violating state of the SND-junction. Finally, we like to emphasize that this effect is only possible in connection with unconventional superconductivity and cannot occur for standard SNS-junctions. Therefore, high temperature superconductivity provides an exciting new class of Josephson phenomena.

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